

JC13 Rec'd PCT/PTO 22 APR 2005

AMENDMENT

(Amendments under Section 11 of the Japanese Law Concerning
International Applications, Etc. Pursuant to
the Patent Cooperation Treaty)

To Mr. Mitsuji UEMAE, Examiner of the Patent Office

1. Identification of International Application
PCT/JP03/13520
2. Applicant
Name: DAIWABO CO., LTD.
Address: 6-8, Kyutaromachi 3-chome, Chuo-ku, Osaka-shi, Osaka
541-0056 JAPAN

Nationality: Japan
Residence: Japan

3. Agent
Name: IKEUCHI SATO & PARTNER PATENT ATTORNEYS
Address: 26th Floor, OAP TOWER, 8-30, Tenmabashi 1-chome,
Kita-ku, Osaka-shi, Osaka 530-6026 JAPAN

4. Items to be Amended
Specification and the claims

5. Subject Matter of the Amendments
As per the attached sheets.

(1) In the specification (page 5, line 21 (corresponding to page 5, lines 21-22 in the English language translation of the originally filed application)), "that gels under heat and humidity to form a gel material" is amended to "that gels under heat and humidity and is pressed and spread by pressing to form a film-like gel material".

(2) In the specification (page 6, line 3 (corresponding to page 5, line 32 in the English language translation of the originally filed application)), "has at least the following steps" is amended to "has at

least all of the following steps A to D".

(3) In the specification (page 6, line 11-13 (corresponding to page 6, lines 2-8 in the English language translation of the originally filed application)), "to a heat-and-humidity treatment (hereinafter referred to as a "gel processing") using a heat treatment device that is set to a certain temperature within a range of no less than a temperature at which the heat-and-humidity gelling resin gels and no more than "the melting point of the heat-and-humidity gelling resin - 20°C", to cause the heat-and-humidity gelling resin to gel, and fixing the other fiber using the heat-and-humidity gelling resin gel" is amended to "to pressing and a heat-and-humidity treatment (hereinafter referred to as a "gel processing") using a heat treatment device that is set to a certain temperature within a range of no less than a temperature at which the heat-and-humidity gelling resin gels and no more than "the melting point of the heat-and-humidity gelling resin - 20°C", to cause the heat-and-humidity gelling resin to gel and be pressed and spread to form a film, and fixing the other fiber using the heat-and-humidity gelling resin gel".

(4) In the claims (page 46, line 3 in claim 1 (corresponding to page 47, lines 8-9 in the English language translation of amended claim 1 under Article 19)), "gel under heat and humidity and be pressed and spread" is amended to "gel under heat and humidity and be pressed and spread by pressing".

6. List of Attached Documents

(1) The specification, pages 5 and 6 (corresponding to pages 4, 5, 6 and 6-1 in the English translation)

(2) The claims, page 46 (corresponding to pages 47, 47-1 and 48, 48-1 in the English translation)

thickness or the like, or irregular pore diameters are likely to occur in the nonwoven. Therefore, the electrolytic solution cannot be kept uniform, or both a fine powder short circuit and a dendritic short circuit are likely to occur, resulting in a high defect rate of a battery (hereinafter also referred to as a "battery defect rate"). When pressure bonding using a thermal roller or the like is performed in order to decrease the pore diameter and thickness of a nonwoven, significant fusion bonding occurs on a surface of the nonwoven (dense surface) and less inside the nonwoven (coarse inside), leading to an increase in the battery defect rate. Further, the electrolytic solution is not kept uniform, so that an internal resistance of the battery is increased. In the separator of Patent Publication 4, a wetlaid nonwoven having a low mass per unit area of 12 to 14 g/m² and a predetermined thickness, which contains a splittable composite fiber, is produced, and thereafter, the wetlaid nonwoven is immersed in an aqueous solution of polyalkylene denatured polysiloxane, thereby attempting to decrease the micropore diameter of the nonwoven. However, for such a low-mass per unit area nonwoven, it is difficult to produce a nonwoven having a uniform mean flow pore diameter and bubble point pore diameter. In fact, the nonwoven has a large variation in pore diameter, leading to instable puncture strength. Further, a splittable composite fiber containing an ethylene-vinyl alcohol copolymer as at least one component is mixed with a hot melt fiber to obtain a wetlaid nonwoven, which is in turn subjected to a dry heat calender process at a processing temperature that causes the hot melt fiber to exhibit its adhesion ability. Therefore, only the hot melt fiber contributes to adhesion ability, so that the puncture strength is insufficient. In the separator of Patent Publication 5, a splittable composite fiber made of two components, i.e., polypropylene/polyester, nylon 66/polyester, and polypropylene/polyethylene, is split into plate-like microfine fibers, which are in turn subjected only to a heat calender process at a temperature that is below the melting point of the lower-melting point component. Therefore, it is difficult to obtain a nonwoven having a uniform mean flow pore diameter and bubble point pore diameter, resulting in a nonwoven having a significant variation in pore diameter. Therefore, no stable puncture strength is acquired. Although Patent Publications 6 to 9 disclose separators containing fibers that are bonded

under heat and humidity, all the separators are intended to be used for an alkaline battery. It is difficult to obtain a separator having a small pore diameter that is required for an organic electrolyte battery.

5 Disclosure of Invention

The present invention is provided to solve the above-described problems. An object of the present invention is to provide an organic electrolyte battery separator made of a nonwoven that can be produced inexpensively, has an excellent yield in production, has an excellent level
10 of electrolytic solution holding ability, and can prevent a fine powder short circuit and a dendritic short circuit when incorporated into a battery (i.e., a low battery defect rate), in place of nonwovens that conventionally have been proposed as organic electrolyte battery separators. Another object of the present invention is to provide an
15 organic electrolyte battery that has an excellent level of safety, has a short circuit less often, and has excellent battery characteristics.

The organic electrolyte battery separator of the present invention is made of a nonwoven containing a resin that can gel by heating in the presence of moisture (hereinafter referred to as a “heat-and-humidity
20 gelling resin”) and another fiber. The other fiber is fixed by the heat-and-humidity gelling resin that gels under heat and humidity and is pressed and spread by pressing to form a film-like gel material (hereinafter referred to as a “gel material”). The nonwoven has a mean flow pore diameter of 0.3 to 5 μm and a bubble point pore diameter of 3
25 to 20 μm as measured in accordance with ASTM F 316 86.

The organic electrolyte battery separator of the present invention can be produced using the following method. Specifically, a method for producing an organic electrolyte battery separator comprising a heat-and-humidity gelling fiber in which a resin capable of gelling by
30 heating in the presence of moisture (hereinafter referred to as a “heat-and-humidity gelling resin”) is present on at least a portion of a surface of the fiber, and another fiber, has at least all of the following steps A to D:

- 35 A. preparing a nonwoven sheet comprising a heat-and-humidity gelling fiber and another fiber;
- B. subjecting the nonwoven sheet to a hydrophilic treatment;
- C. providing moisture to the hydrophilic-treated nonwoven sheet

(hereinafter referred to as a "hydrophilic nonwoven sheet") to obtain a water-containing sheet; and

5 D. subjecting the water-containing sheet to pressing and a heat-and-humidity treatment (hereinafter referred to as a "gel processing") using a heat treatment device that is set to a certain temperature within a range of no less than a temperature at which the heat-and-humidity gelling resin gels and no more than "the melting point of the heat-and-humidity gelling resin - 20°C", to cause the heat-and-humidity gelling resin to gel and be pressed and spread to form
10 a film, and fixing the other fiber using the heat-and-humidity gelling resin gel.

An organic electrolyte battery of the present invention is obtained by incorporating the separator.

15 Brief Description of Drawings

FIG. 1 is a cross-sectional view showing a method of measuring a contact angle on a surface of a nonwoven used in an example of the present invention.

20 FIG. 2 is a 200x SEM microscopic photograph of a surface of a nonwoven sheet obtained in Example 1 of the present invention.

FIGS. 3A to 3D are 200x SEM microscopic photographs of a surface of a battery separator obtained in Example 1 of the present invention.

25 FIG. 4 is a 200x SEM microscopic photograph of a surface of the battery separator obtained in Example 1 of the present invention.

30 FIGS. 5A and 5B are 300x SEM microscopic photographs of a surface of a nonwoven sheet obtained in Example 5 of the present invention. FIGS. 5C and 5D are 300x cross-sectional photographs of a surface of the nonwoven sheet obtained in Example 5 of the present invention.

FIGS. 6A and 6B are 300x SEM micrographs of a surface of a battery separator obtained in Example 5 of the present invention. FIGS. 6C and 6D are 1000x cross-sectional photographs of a surface of the battery separator obtained in Example 5 of the present invention.

35 1: glass plate, 2: sample, 3: pure water

Best Mode for Carrying Out the Invention

The present inventors have diligently researched to conceive that a separator made of a nonwoven that excellently resists a fine powder short circuit can be obtained by establishing an appropriate mean flow pore diameter range and bubble point pore diameter range, but not
5 sufficiently by only decreasing a pore diameter. It was found that this

CLAIMS

1. (Amended) An organic electrolyte battery separator, which is composed of a nonwoven comprising a heat-and-humidity gelling resin capable of gelling by heating in the presence of moisture and another fiber,
5 the other fiber being fixed with a film-like gel material obtained by causing the heat-and-humidity gelling resin to gel under heat and humidity and be pressed and spread by pressing, and
10 the nonwoven having a mean flow pore diameter of 0.3 to 5 μm and a bubble point pore diameter of 3 to 20 μm as measured in accordance with ASTM F 316 86.
2. The organic electrolyte battery separator according to claim 1,
15 wherein the heat-and-humidity gelling resin is a heat-and-humidity gelling fiber, the heat-and-humidity gelling resin being provided at least at a portion of a surface of the heat-and-humidity gelling fiber.
3. The organic electrolyte battery separator according to claim 1,
20 wherein a proportion of the nonwoven occupied by the heat-and-humidity gelling resin is in a range of 10 to 50 mass%.
4. The organic electrolyte battery separator according to claim 1,
25 wherein the heat-and-humidity gelling resin is an ethylene-vinyl alcohol copolymer.
5. The organic electrolyte battery separator according to claim 1,
wherein the other fiber has a fiber diameter of 15 μm or less.
6. The organic electrolyte battery separator according to claim 1,
30 wherein an average fiber diameter of the other fiber constituting the nonwoven is 10 μm or less.
7. The organic electrolyte battery separator according to claim 1,
35 wherein the fiber constituting the nonwoven other than the heat-and-humidity gelling resin is an olefin fiber.

8. The organic electrolyte battery separator according to claim 1,

wherein the other fiber includes a high-strength fiber having a single fiber strength of 4.5 cN/dtex or more in a range of 5 to 250 parts by mass where the heat-and-humidity gelling resin is assumed to be 100 parts by mass.

5

9. The organic electrolyte battery separator according to claim 1, wherein the other fiber includes a heat-melting fiber that does not substantially shrink at a temperature that causes the heat-and-humidity gelling resin to gel under heat and humidity to fix the other fiber, in a
10 range of 10 to 300 parts by mass where the heat-and-humidity gelling resin is assumed to be 100 parts by mass.

10. The organic electrolyte battery separator according to claim 1, wherein the nonwoven further comprises a synthetic pulp in addition to
15 the other fiber.

11. The organic electrolyte battery separator according to claim 1, wherein the synthetic pulp is included in a range of 10 to 200 parts by mass where the heat-and-humidity gelling resin is assumed to be 100
20 parts by mass.

12. The organic electrolyte battery separator according to claim 2, wherein an average fiber diameter of the heat-and-humidity gelling fiber and the other fiber is 10 μm or less.
25

13. The organic electrolyte battery separator according to claim 2, wherein the heat-and-humidity gelling fiber has a fiber diameter of 1 to 6 μm .

30 14. The organic electrolyte battery separator according to claim 13, wherein the heat-and-humidity gelling fiber is a fiber provided by splitting a splittable composite fiber that contains the heat-and-humidity gelling resin and another resin, which are adjacent to each other in a cross-section of the fiber.

15. The organic electrolyte battery separator according to claim 14, wherein, when the splittable composite fiber comprised of the